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Recent high resolution measurements of the momenta of two electrons emitted from Argon atoms in a strong laser field show a strong preference for the outgoing electrons to have similar momenta and to be ejected in the same direction along the field. We discuss the final stages of this process in a classical model in the spirit of Wannier's approach to double ionization after electron impact. Starting from an intermediate state of high but negative energy the field opens up a channel through which both electrons can escape. Near the threshold for this process Coulomb repulsion favors an escape with the electron momenta and positions symmetric with respect to the electric field axis. Classical trajectory simulations within this symmetry subspace account for most features of the observed momentum distribution.

Double ionization of neutral atoms (in particular He, Ne and Ar) in the presence of strong fields has attracted considerable attention after it was noted that the observed yield is much higher than can be expected on the basis of an independent electron model [1,2]. It was proposed early on that correlations between the electrons should be responsible for this enhancement. A striking demonstration of such correlations is provided by recent high resolution experiments on the distribution of the ion-recoil momenta [3,4,6] and the electron momenta [5] which clearly show a preference for the symmetric escape of both electrons to the same side of the nucleus. This is very different from the process in the absence of a field (Wannier-ionization, [7,8]) where the dominant ionization path has both electrons escape symmetrically placed on opposite sides of the nucleus. In particular, while the Wannier mode minimizes the electron repulsion, a symmetric escape to the same side has considerable residual energy in the repulsion between electrons. So why is this channel favoured?

It is by now generally accepted that the ionization takes place in two steps: a high excitation of one electron followed by the double ionization event [9–16]. The highly excited electron is accelerated by the field and driven back to the core where it collides with the other electron, and transfers enough energy and momentum so that both electrons can escape from the nucleus. It is this rescattering of the electron that enhances the energy transfer and thus the ionization rate. But it also implies that during the ionization process the interaction between the electrons cannot be ignored.

The amount of energy the excited electron can gain during half a period of the electric field has been estimated in [3]: for a field intensity of  $2.9 \cdot 10^{14} \text{ W/cm}^2$  the estimate gives a maximal energy transfer of about 60 eV. This is insufficient to ionize the second electron directly, but as the experiments show, double ionization is possible nevertheless. This implies that there must be a mechanism by which the electrons can draw additional energy

from the field so that asymptotically, once the pulse has ceased, the total energy of the system is positive.

The related problem of double ionization in collisions or single photon excitation was studied in a classic paper by Wannier [7,8]. He assumed that during the first stages a high energy complex of electrons close to the nucleus is formed from which then the ionized electrons escape. If the energy is close to threshold, they cannot afford to put energy into the mutual repulsion and the escape is with both electrons on opposite sides of the nucleus. Moreover, he argued that their distance has to be the same, for any difference in position and energy would be amplified, pushing the configuration towards single electron ionization.

Essential elements of this discussion also apply to the present situation. At the end of the first step the rescattering of one electron to the core produces a highly excited two electron complex. The estimates show [3] that the total energy in the system is insufficient to double ionize immediately. However, if the electric field during the collision is non-zero, a saddle opens through which the electrons can escape. Because of their mutual repulsion the electron that gets to the saddle first has an advantage: it can cross the saddle while pushing the other back to the nucleus. Since energy is scarce near threshold this results in either single ionization or in another rescattering event, but most likely not in double ionization. This suggests that the dominant path leading to double ionization has both electrons cross the saddle side by side. This singular process then acquires a finite probability when trajectories asymptotic to this configuration are taken into account.

Therefore, we propose that near the threshold for double ionization the only path leading to double ionization has both electrons escape symmetric with respect to the field axis. With the field pointing along the  $x$ -axis and the electrons confined to the plane  $z = 0$  their coordinates are  $(x, y, 0)$  and  $(x, -y, 0)$  in position and  $(p_x, p_y, 0)$  and  $(p_x, -p_y, 0)$  in momenta. In this geometry the obser-

vation that both electrons escape to the same side is built from the outset. As an aside we note that this symmetry plane also contains the Wannier orbit, for  $x = 0$ , as the symmetric escape perpendicular to the field axis.

The classical Hamilton function for this geometry then is (in atomic units, with infinitely heavy nucleus and in dipole approximation)

$$H(p_x, p_y, x, y, t) = p_x^2 + p_y^2 + V(x, y, t) \quad (1)$$

with potential energy

$$V(x, y, t) = -\frac{4}{\sqrt{x^2 + y^2}} + \frac{1}{2y} + 2F x f(t) \cos(\omega t + \phi) \quad (2)$$

and the pulse shape

$$f(t) = \sin^2(\pi t/T_d) \quad (3)$$

where the duration of the pulse is taken to be four field cycles,  $T_d = 8\pi/\omega$ . The frequency is  $\omega/2 = 0.057 \text{ a.u.}$ , and corresponds to the experimental situation [3–5]. The rescattering of the electrons leads to a highly excited complex of total energy  $\tilde{E}$  which every now and then is close to the symmetric configuration described by the Hamiltonian (1). Any configuration on this energy shell (for some fixed time  $t$ ) as well as any phase  $\phi$  of the field is equally likely, and the experimental observations are averages over initial conditions and phases.

As mentioned, for the weakest fields where double ionization is observed the rescattered electron does not bring in enough energy for double ionization. However, if the collision happens near a time where the field is strong, the electric field distorts the potential and opens a path for escape in down-field direction. This process can be discussed adiabatically for fixed external field since the motion of the electrons near the nucleus is much faster than the change in the field. The ionization can thus be discussed in the potential (2) with fixed field. Equipotential lines for the potential (2) at a maximum of the field for  $F = 0.137 \text{ a.u.}$ , corresponding to an intensity of  $6.6 \cdot 10^{14} \text{ W/cm}^2$ , are shown in Fig. 1. The saddle is located along the line  $x = r_S \cos \theta$  and  $y = r_S \sin \theta$  with  $\theta = \pi/6$  or  $5\pi/6$  and at a distance  $r_S^2 = \sqrt{3}/F_{max}$  where  $F_{max} = \max_t |F f(t) \cos(\omega t + \phi)|$ . The energy of the saddle is

$$V_S = -6\sqrt{F_{max}/\sqrt{3}}. \quad (4)$$

For the extremal fields in a pulse of the above mentioned intensity this gives  $V_S = -1.69 \text{ a.u.}$ , so that within the adiabatic picture the saddle can be reached if the returning electron brings in at least  $1.22 \text{ a.u.}$  in energy. During a field cycle the saddle moves in from infinity along the line at  $\theta = \pi/6$ , moves out again to infinity after half a period and then moves in and out again along the line  $\theta = 5\pi/6$  during the second half of the cycle. Ionization is most likely when the saddle is closest to the nucleus.

A typical trajectory within the symmetric configuration is shown in Fig. 2. During the ramping of the field the electronic motion is little influenced by the electric field, but during the third half cycle of the field the saddle is close enough to the electron orbits and ionization takes place. Once on the other side of the saddle, the electrons rapidly gain energy. The saddle thus provides a kind of transition state [17,18] for the double ionization process: once the electrons cross it, they are accelerated by the field and pulled further away, making a return rather unlikely. Moreover, they can acquire the missing energy so that both electrons can escape even when the field vanishes. The field thus plays a double role in determining a threshold for this process: during the first stages of the rescattering process it provides the energy for the collision complex and during the final stages it opens the path for double escape.

In the experiments it is not possible to monitor all details and intermediate stages of the process. Most information is extracted from the distribution of final momenta  $\mathbf{p}_n$  of the nucleus and  $\mathbf{p}_i$  of the electrons, where  $\mathbf{p}_1 + \mathbf{p}_2 \approx -\mathbf{p}_n$  [3–6]. Because of the symmetry assumption in the model the components perpendicular to the field vanish. The distribution of the parallel components can be calculated by averaging over all initial conditions of prescribed energy and all phases of the field.

Classical scaling of the Hamiltonian (1) implies that the results do not depend on the initial energy  $\tilde{E}$  and field strength independently, but on the combination  $F/\tilde{E}^2$  only. The field strength  $F$  is set by the intensity of the laser. The initial energy of the two-electron complex is determined by the field dependent efficiency of the single electron excitation step and thus not directly accessible, although it can be estimated as in [3]. We therefore fix  $F$  and vary initial energy. The results for fixed field strength  $F = 0.137 \text{ a.u.}$  and an initial energy of  $\tilde{E} = -0.58 \text{ a.u.}$  are compared to the experimental distribution in Fig. 3. The final distribution of momenta clearly shows the double hump structure indicating a preference for ionization parallel and antiparallel to the field. The maxima of the experimental distribution are at about  $p_{\parallel} = \pm 1.5 \text{ a.u.}$ , whereas the numerical ones within the symmetric subspace lie at about  $\pm 1 \text{ a.u.}$ . We take this close agreement as strong indication that double ionization can only occur in the neighborhood of the symmetric process discussed here.

For lower energy,  $\tilde{E} = -1.3 \text{ a.u.}$  the minimum at  $p_{\parallel} = 0$  almost vanishes and only a single maximum shows up (Fig. 4a). This corresponds to the experimental situation of a weaker pulse which evidently transfers less energy to the rescattered electron [3]. Within the symmetric subspace, there are two reasons for this change: at this lower initial energy the electrons have little kinetic energy when crossing the barrier, so that the splitting should be expected to be small. Secondly, the electrons cross the barrier typically when the field is strongest, so that after the ionization there is still considerable smearing of the

distribution due to the interaction with the remainder of the pulse. If the distribution is monitored immediately after the crossing of the barrier the two preferred momenta parallel and anti-parallel to the field stand out clearly, as demonstrated in Fig. 4b.

These numerical results in the reduced symmetry subspace are in surprising agreement with the experimental data. They show that the configuration with rotational symmetry around the field axis dominates the cross section. The interaction with the field shortly after ionization is responsible for most of the smearing of the final distribution, additional contributions come from trajectories that are not symmetric. Actually, non-symmetric configurations are needed in order to obtain a non-vanishing cross section to begin with: the symmetric subspace is a set of measure zero in the phase space of the six degree of freedom system and acquires a finite overlap with initial conditions only due to non-symmetric initial conditions that are asymptotic to the symmetric subspace. The cross section for double ionization shows that this overlap is small [3–6].

The picture proposed here for multiphoton double ionization is very similar to that of Wannier for double ionization through electron impact. The main difference is that now the symmetry between the outgoing electrons is a rotation around the field axis whereas it is a point symmetry with the nucleus in the center in Wanniers case.

Finally, we would like to mention that besides the different symmetry there is another difference to the Wannier double ionization without field: In the zero field ionization there is only a single trajectory so that there in leading order semiclassical approximation no quantum interferences can be expected [19]. The present problem falls into the category of quantum chaotic scattering [20,21] where the classical ionization dynamics is chaotic and quantum interference effects between different paths cannot be ruled out. It will be interesting to pinpoint quantum interference effects in this system.

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FIG. 1. Adiabatic potential  $V(x, y, t)$  for fixed time  $t$  in the symmetric subspace. The saddle moves along the dashed line when the electric field points in the positive  $x$ -direction and along a second obtained by reflection on  $x = 0$  during the other half of the field cycle.

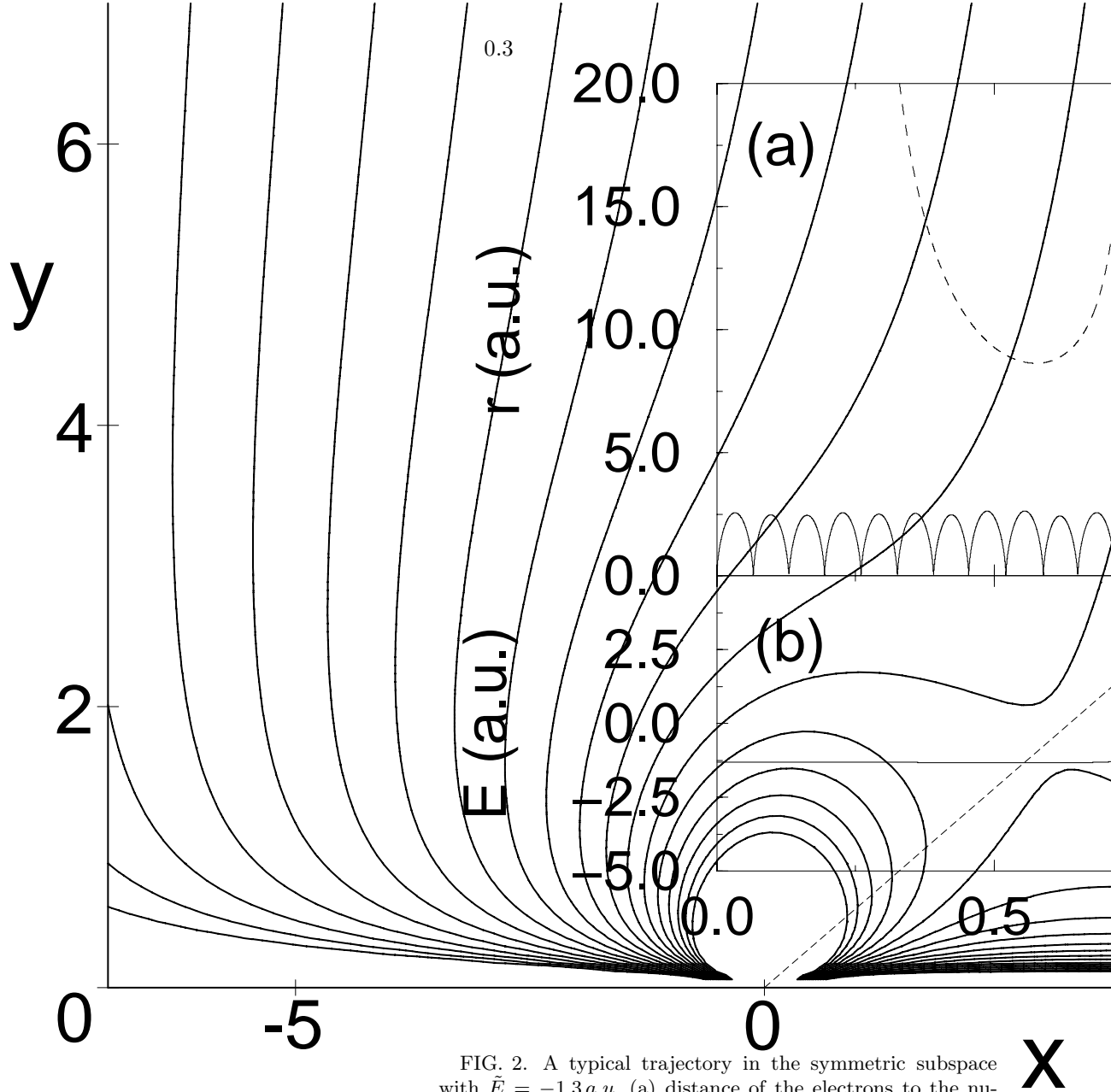


FIG. 2. A typical trajectory in the symmetric subspace with  $\bar{E} = -1.3 a.u.$  (a) distance of the electrons to the nucleus. The dashed line indicates the distance of the saddle. Note that before the double ionization occurs the effect of the field on the electrons is minimal, supporting the adiabatic assumption. (b) energy of the electrons. Note that the initial state has negative total energy and cannot lead to double ionization. The energy increases once the electrons have escaped from the nucleus far enough so that acceleration by the electric field dominates.

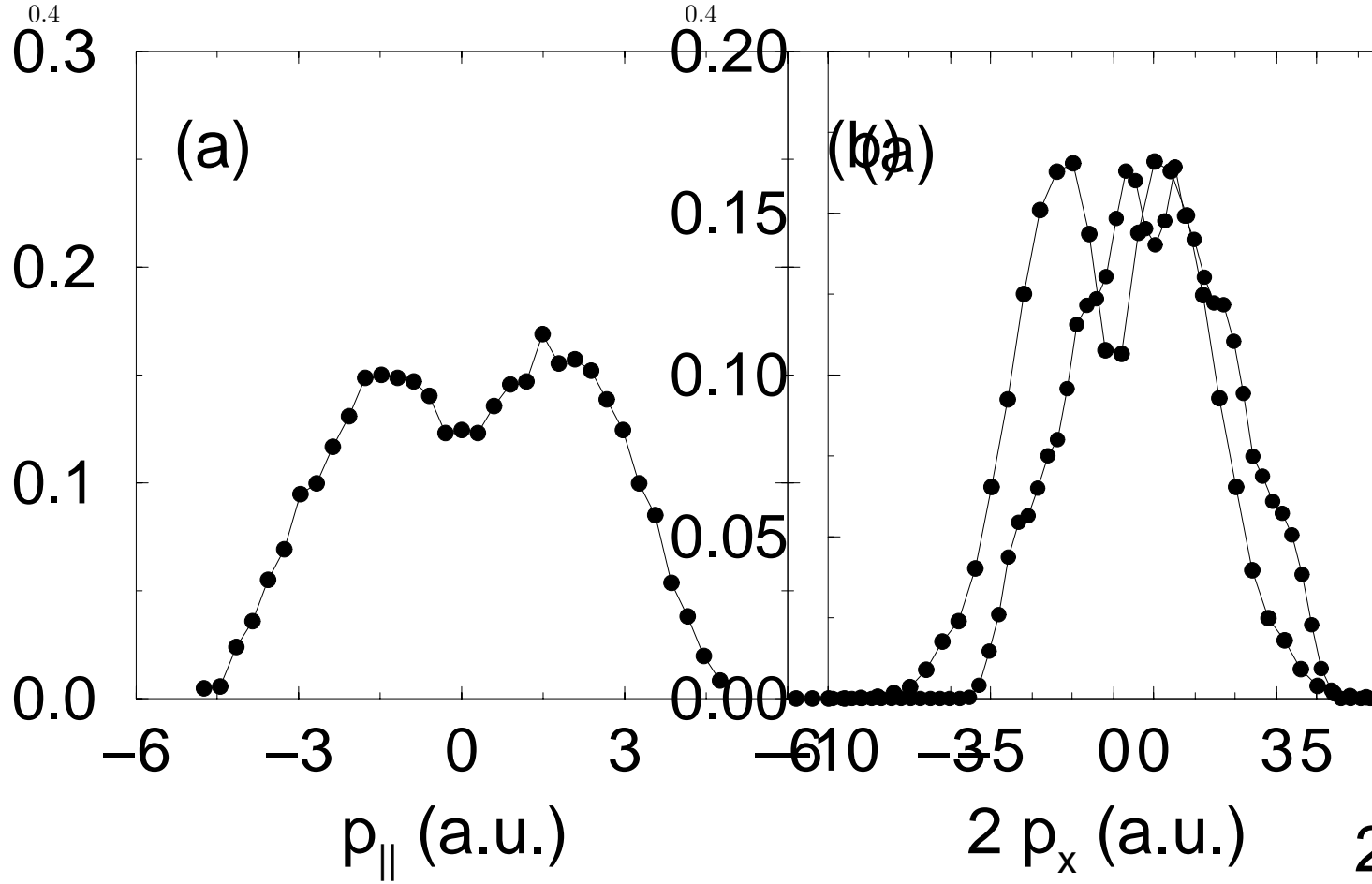


FIG. 3. Final distribution of ion momenta parallel to the field for  $F = 0.136$  a.u.: (a) experimental distribution from [3] (b) distribution from symmetric subspace with initial energy  $\tilde{E} = -0.58$  a.u.. The classical distribution is based on an ensemble of  $2 \cdot 10^5$  trajectories.

FIG. 4. Final distribution of momenta parallel to the field for  $F = 0.136$  a.u. and initial energy  $\tilde{E} = -1.3$  a.u.. (a) at the end of the pulse; (b) at the first zero of the field after crossing the saddle. This figure demonstrates the smearing of the distribution due to the final post ionization interaction with the electric field. It is based on 50,000 classical trajectories.

